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THEORY OF DIFFUSION OF RADON AND THORON THROUGH MEMBRANES UNDER STEADY-STATE AND TIME-DEPENDENT RADIATION CONDITIONS - PART II (TIME-DEPENDENT CASE)

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THEORY OF DIFFUSION OF RADON AND THORON THROUGH MEMBRANES UNDER STEADY-STATE AND TIME-DEPENDENT CONDITIONS - PART II. (TIME-DEPENDENT CASE)

by

J. Bigu*

ABSTRACT

The diffusion of radon and thoron through a membrane separating a well-defined region of space (sensitive volume) from the rest (outside volume) is discussed. Expressions are provided for the direct diffusion process and the reverse diffusion process. In addition, the growth and decay of the radon and thoron progeny formed by the decay of the diffusing radon and thoron, respectively, are discussed.

The cases presented here apply specifically to time-dependent conditions, whereby time-dependent condition is to be understood as the case where the radioactive gas concentration in the outside volume varies with time in a predictable fashion. Three time-dependent cases of practical interest have been chosen, namely those in which the gas concentration in the outside volume is assumed to vary sinusoidally, linearly and exponentially. The data presented here are relevant for theoretical purposes and for practical applications including personal dosimetry and environmental monitoring.

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THÉORIE DE LA DIFFUSION DU RADON ET DU THORON À TRAVERS LES MEMBRANES EN RÉGIME PERMANENT ET EN FONCTION DU TEMPS PARTIE II. (CAS - DIFFUSION EN FONCTION DU TEMPS)

par

J. Bigu*

RÉSUMÉ

Le rapport porte sur la diffusion du radon et du thoron à travers la membrane qui sépare un espace bien défini (volume sensible) du reste (volume extérieur). Il présente des formules relatives aux processus de diffusion directe et inverse. De plus, la vitesse de désintégration des produits de filiation du radon et du thoron, respectivement, activée par la désintégration du radon et du thoron qui se diffusent, est examinée.

Les exemples dont il est question ici s'appliquent en particulier dans les cas de diffusion en fonction du temps, c'est-à-dire, quand la concentration radioactive de gaz dans le volume extérieur varie de façon prévisible avec le temps. On a choisi trois exemples d'ordre pratique qui illustrent ce fait, à savoir, des cas dans lesquels la concentration de gaz dans le volume extérieur semble varier de façon sinusoïdale, exponentielle et linéaire. Les données présentées dans ce rapport se prêtent à des fins théoriques et à des applications pratiques, y compris le contrôle des conditions environnementales.

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INTRODUCTION

The ability to separate materials from each other is an interesting and useful property of certain organic materials usually referred to as membranes. The separation of gases and/or airborne particulates is of particular interest in the radiation field. Radon, thoron and their decay products are known to permeate through membranes of organic materials in varying degrees.

An interesting feature of using organic membranes for separating radioactive species lies in the fact that product separation is accomplished, not only by the different diffusion rate of the diffusing species, but also because of the different radioactive half-lives of the species. Hence, if the rate of diffusion of a given radioactive product is so slow that the time that it would take for the product to diffuse through the membrane by purely diffusional processes is significantly larger than its half-life, virtually complete removal of the product will be realized on the basis of radioactive decay alone during the diffusion process. However, it should be noted that it is quite possible for the daughter products formed from decay of the parent to reach the 'other side' of the membrane, and hence, diffuse through.

Separation of radioactive species based on the combined effect of diffusion and radioactive decay during the diffusion process constitutes a remarkably useful feature of certain organic membranes. The general properties of organic membranes to separate gases, to separate radon and thoron from their decay products, and to separate radon from thoron, has been investigated by several authors (1-6). Some practical applications have been found for these properties.

In a previous paper (Part I, ref. 7), the theory of diffusion of radon and thoron through a membrane separating a well-defined region of space, i.e., 'sensitive' volume, from the rest (outside volume) was investigated. In the

above work, steady-state conditions were assumed, i.e., the radioactive gas concentration remained constant during the diffusion process. In the present paper, diffusion of radioactive gas through a membrane under time-dependent conditions is studied. It should be noted that 'under time-dependent conditions' is to be understood as the case where the radioactive gas concentration in the outside volume (region) varies with time in a predictable fashion. As in the previous paper (7), analytical expressions are provided suitable for personal dosimetry and environmental monitoring, and for other practical and theoretical purposes.

THEORETICAL BACKGROUND

The following items of interest are dealt with below:

1. Diffusion of radon and/or thoron into a sensitive volume, initially free of radioactive gas. This process will be referred to in the future as the direct diffusion process;
2. Diffusion of radon and/or thoron out of the sensitive volume into a region with negligible radon and/or thoron concentration, i.e., radioactive gas-free. This process will be referred to in the future as the reverse diffusion process; and
3. Calculation of radon daughter concentration and/or thoron daughter concentration formed in the direct and reverse diffusion processes.

In order to simplify items 1 to 3, the following assumptions have to be made:

1. Radon daughters and thoron daughters are essentially removed, i.e., filtered out by the membrane. This is true of any membrane, except for very high porosity media of little interest in the context of this paper.
2. Back-diffusion of radioactive gas is negligible during both the direct and reverse diffusion processes.

With the geometry sketched in Fig. 1 with a source of radon or thoron of concentration N_1 in volume V_1, i.e., outside region, separated from the sensitive volume, V_2, by a membrane of thickness δ, surface area A and permeability k, one may write the kinetic equations describing the diffusion process as follows:

$$\frac{dN_1}{dt} = - \lambda_n N_1 + a - \frac{QN_1}{V_1} - \frac{kA}{\delta V_1} (N_1 - N_2) \qquad \text{Eq 1}$$

and

$$\frac{dN_2}{dt} = - \lambda_n N_2 + \frac{kA}{\delta V_2} (N_1 - N_2) \qquad \text{Eq 2}$$

where a is the production rate of the source (radon or thoron). The symbol λ_n stands for the radioactive decay constant of the gas (radon or thoron) and Q is the air flow in the outside region. It should be noted that the first, third and fourth terms on the right hand side of Eq. 1 describe, respectively, the removal of the radioactive gas in volume V_1 by radioactive decay, by forced ventilation or air flow, and by diffusion through the membrane. The first and second terms on the right hand side of Eq. 2 describe, respectively, the removal of radioactive gas in V_2 by radioactive decay and the increase in gas concentration in the same volume by diffusion through the membrane from V_1.

Equations 1 and 2 can be rearranged as follows:

$$\frac{dN_1}{dt} + MN_1 = a + SN_2 \qquad \text{Eq 3}$$

and

$$\frac{dN_2}{dt} + PN_2 = TN_1 \qquad \text{Eq 4}$$

where,

$$M = \Lambda + \frac{kA}{\delta V_1}, \quad \Lambda = \lambda_n + Q/V_1$$

$$S = \frac{kA}{\delta V_1}, \; P = \lambda_n + \frac{kA}{\delta V_2} \text{ and } T = \frac{kA}{\delta V_2}$$

The solutions to Equations 3 and 4 for the steady-state case were

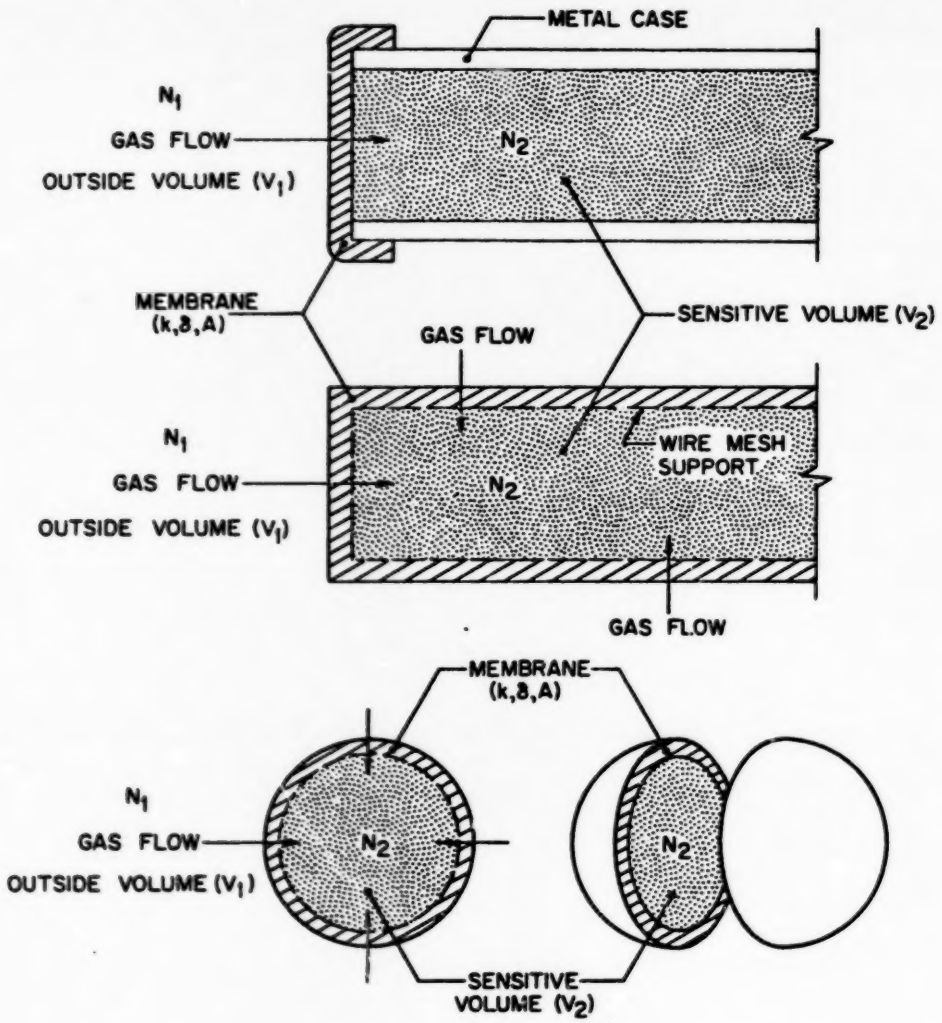

Fig. 1 - Outside volume/diffusion membrane/sensitive volume arrangement for three different experimental configurations.

obtained by the conditions $dN_1/dt=0$, i.e., N_1 is a constant (7). For the time-dependent case, N_1 may vary in any arbitrary fashion by changing a and/or Q accordingly, see Equation 1. However, in this paper Equation 4 is solved for specific time-dependent analytical expressions that describe the dynamic behaviour of N_1. A more rigorous treatment in which N_1 in Equation 3 is derived for specific time-dependent situations for a and/or Q, is dealt with elsewhere (8).

Equation 4 has been solved by writing:

$$N_1 = N_1^0 F(t) \qquad \text{Eq 5}$$

where, F(t) can assume the following forms:

$$F(t) = 1 + a' \sin\omega t \quad \text{(sinusoidal case)} \qquad \text{Eq 6}$$

$$F(t) = 1 + \eta t \quad \text{(linear case)} \qquad \text{Eq 7}$$

$$F(t) = e^{\delta t} \quad \text{(exponential case)} \qquad \text{Eq 8}$$

$$F(t) = F_0 \text{ for } t \leq t_0, \text{ and}$$

$$F(t) = qF_0 \text{ for } t > t \qquad \text{Eq 9}$$

where, $q \neq 1$, but $q \gtrless 0$

The symbols a', η, δ and q are the 'perturbation' amplitudes corresponding to Equations 6, 7, 8 and 9, respectively. For a'=0, η=0, and δ=0, $F(t)$=1, i.e., non-perturbed, or steady-state case.

The radon progeny and thoron progeny formed in V_2 upon decay of radon and thoron, respectively, diffusing through the membrane are calculated according to the following differential equation which describes the growth of decay products from their original parent product:

$$\frac{dN_i}{dt} = N_{i-1}\lambda_{i-1} - N_i\lambda_i \qquad \text{Eq 10}$$

where the index i in Equation 10 refers to RaA, RaB, RaC for radon daughters and ThA, ThB, ThC and ThC' for thoron daughters. The index i-1 refers to the parent product of daughter product i, i.e., Rn-222 for RaA, and so on. N_i in

Equation 10 can be solved according to standard methods of differential calculus. Notice that for i≡RaA or ThA, $N_{i-1}=N_2$ which is given by Equations 15, 43 and 66, below. It should also be noted that the solution for N_i according to Equation 10, is then used to solve the differential equation describing the growth of its daughter product, and so on. Hence, the calculation increases considerably in complexity for subsequent daughter products.

The activity, A, corresponding to the radioactive gas and its decay products is given by the well-known relationships:

$$A_n = N_n\lambda_n \quad \text{and} \quad A_i = N_i\lambda_i \qquad \text{Eq 11}$$

where, for simplicity, consistency and generality, the index n is used for the radioactive gas in V_2 instead of the index 2 of Equations 1 to 4.

1. DIRECT DIFFUSION PROCESS

Three time-dependent cases of practical interest are discussed here, namely: sinusoidal, linear and exponential. The step-function case described by Equation 9 has been dealt with elsewhere (7,9).

The atom concentration and activity in V_2, corresponding to the radioactive gas and its progeny, can be obtained by means of Equations 3, 4, 10 and 11 with Equation 5 and the following boundary conditions: $N_1(t=0) = N_1^0$, $N_2(t=0) = N_n(t=0) = 0$, and $N_i(t=0) = 0$. The integrated activity during the direct diffusion process, i.e., sampling period, IAS, for the radioactive gas and its progeny is given, respectively, by Equations 12 and 13:

$$IAS(n) = \int_{T_1}^{T_2} N_n\lambda_n dt \qquad \text{Eq 12}$$

$$IAS(i) = \int_{T_1}^{T_2} N_i\lambda_i dt \qquad \text{Eq 13}$$

where, T_1 and T_2 are the limits of integration with $T_1 \geq 0$ and $T_2 \geq T_1$. Hence, T_2-T_1 is the sampling time. Usually T_1 is taken as the beginning of sampling, i.e., $T_1=0$.

1a. SINUSOIDAL CASE

For this case, Equation 5 becomes:

$$N_1 = N_1^0 (1 + a'\sin\omega t) \qquad \text{Eq 14}$$

which, upon substitution into Equation 4, taking into account the proper boundary condition, gives:

$$N_n = N_1^0 T\left[\frac{1}{P} + \frac{a'(P\sin\omega t - \omega\cos\omega t)}{\omega^2 + P^2} - \left(\frac{1}{P} - \frac{a'\omega}{\omega^2 + P^2}\right)e^{-Pt}\right] \qquad \text{Eq 15}$$

And for the decay products:

$$N_A = N_1^0 T\lambda_n\left[\frac{1}{\lambda_A P} + C\sin\omega t - B\cos\omega t - A'e^{-Pt} - Xe^{-\lambda_A t}\right] \qquad \text{Eq 16}$$

where,

$$A' = \left(\frac{1}{P} - \frac{a'\omega}{\omega^2 + P^2}\right)\left(\frac{1}{\lambda_A - P}\right) \qquad \text{Eq 17}$$

$$B = \frac{a'\omega(P + \lambda_A)}{(\omega^2 + P^2)(\omega^2 + \lambda_A^2)} \qquad \text{Eq 18}$$

$$C = \frac{a'(P\lambda_A - \omega^2)}{(\omega^2 + P^2)(\omega^2 + \lambda_A^2)} \qquad \text{Eq 19}$$

$$X = \frac{1}{\lambda_A P} - (A' + B) \qquad \text{Eq 20}$$

$$N_B = N_1^0 T\lambda_n\lambda_A\left[\frac{1}{\lambda_A\lambda_B P} + C'\sin\omega t - B'\cos\omega t - \frac{A'}{\lambda_B - P}e^{-Pt} - \frac{X}{\lambda_B - \lambda_A}e^{-\lambda_A t} - X'e^{-\lambda_B t}\right] \qquad \text{Eq 21}$$

where,

$$B' = \frac{C\omega + B\lambda_B}{\lambda_B^2 + \omega^2} \qquad \text{Eq 22}$$

$$C' = \frac{C\lambda_B - B\omega}{\lambda_B^2 + \omega^2} \qquad \text{Eq 23}$$

$$X' = \left(\frac{1}{\lambda_A\lambda_B P} - B' - \frac{A'}{\lambda_B - P} - \frac{X}{\lambda_B - \lambda_A}\right) \qquad \text{Eq 24}$$

$$N_C = N_1^0 T\lambda_n\lambda_A\lambda_B\left[D + Y\sin\omega t - Z\cos\omega t - E'e^{-Pt} - Fe^{-\lambda_A t} - Ge^{-\lambda_B t} - X''e^{-\lambda_C t}\right] \qquad \text{Eq 25}$$

where,

$$D = \frac{1}{\lambda_A\lambda_B\lambda_C P} \qquad \text{Eq 26}$$

$$E' = \frac{A'}{(\lambda_B - P)(\lambda_C - P)} \qquad \text{Eq 27}$$

$$F = \frac{X}{(\lambda_B - \lambda_A)(\lambda_C - \lambda_A)} \qquad \text{Eq 28}$$

$$G = \frac{X'}{\lambda_C - \lambda_B} \qquad \text{Eq 29}$$

$$Y = \frac{C'\lambda_C - B'\omega}{\lambda_C^2 + \omega^2} \qquad \text{Eq 30}$$

$$Z = \frac{C'\omega + B'\lambda_C}{\lambda_C^2 + \omega^2} \qquad \text{Eq 31}$$

$$X'' = D - (E' + F + G + Z) \qquad \text{Eq 32}$$

$$N_C' = 0.663\, N_C(\lambda_C/\lambda_C') \qquad \text{Eq 33}$$

The integrated activities are calculated using Equations 12 and 13, and Equations 15, 16, 21 and 25. Hence:

$$IAS(n) = N_1^0 T\lambda_n\left[\frac{T_2 - T_1}{P} + Hf(\sin\omega t) - If(\cos\omega t) + \frac{J}{P}(e^{-PT_2} - e^{-PT_1})\right] \qquad \text{Eq 34}$$

where,

$$H = \frac{a'P}{\omega^2 + P^2} \quad ; \quad I = \frac{a'\omega}{\omega^2 + P^2} \quad ; \quad J = \frac{1}{P} - \frac{a'\omega}{\omega^2 + P^2} \qquad \text{Eq 35}$$

$$f(\sin\omega t) = \int_{T_1}^{T_2} \sin\omega t dt = -\frac{1}{\omega}(\cos\omega T_2 - \cos\omega T_1) \qquad \text{Eq 36}$$

$$f(\cos\omega t) = \int_{T_1}^{T_2} \cos\omega t dt = \frac{1}{\omega}(\sin\omega T_2 - \sin\omega T_1) \qquad \text{Eq 37}$$

Similarly,

$$IAS(A) = N_1^0 T\lambda_n\lambda_A\left[\frac{T_2 - T_1}{\lambda_A P} + Cf(\sin\omega t) - Bf(\cos\omega t) + \frac{A'}{P}(e^{-PT_2} - e^{-PT_1}) + \frac{X}{\lambda_A}(e^{-\lambda_A T_2} - e^{-\lambda_A T_1})\right] \qquad \text{Eq 38}$$

$$IAS(B) = N_1^0 T\lambda_n\lambda_A\lambda_B\left[\frac{T_2 - T_1}{\lambda_A\lambda_B P} + C'f(\sin\omega t) - B'f(\cos\omega t) + \frac{A'}{P(\lambda_B - P)}(e^{-PT_2} - e^{-PT_1}) + \frac{X}{\lambda_A(\lambda_B - \lambda_A)}(e^{-\lambda_A T_2} - e^{-\lambda_A T_1}) + \frac{X'}{\lambda_B}(e^{-\lambda_B T_2} - e^{-\lambda_B T_1})\right] \qquad \text{Eq 39}$$

$$IAS(C) = N_1^0 T\lambda_n\lambda_A\lambda_B\lambda_C\Big[D(T_2 - T_1) + Yf(\sin\omega t) - Zf(\cos\omega t) + \frac{E'}{P}\left(e^{-PT_2} - e^{-PT_1}\right) + \frac{F}{\lambda_A}\left(e^{-\lambda_A T_2} - e^{-\lambda_A T_1}\right) + \frac{G}{\lambda_B}\left(e^{-\lambda_B T_2} - e^{-\lambda_B T_1}\right) + \frac{X''}{\lambda_C}\left(e^{-\lambda_C T_2} - e^{-\lambda_C T_1}\right)\Big] \quad \text{Eq 40}$$

and, $IAS(C') = 0.663\ IAS(C)$. Eq 41

It should be noted in what follows that because of the large number of constants defined in the text and the limited number of letters (symbols) available, the same symbol(s) have sometimes been used to define constants which apply to two different, and hence, unrelated cases, e.g., sinusoidal and linear cases. The reader should be aware of this and exercise special care to use the proper set of constants for the specific case they are meant to apply to.

1b. LINEAR CASE

For this case, Equation 5 becomes:

$$N_1 = N_1^0(1 + \eta t) \quad \text{Eq 42}$$

which, upon substitution into Equation 4, taking into account the proper boundary condition, gives:

$$N_n = \frac{N_1^0 T}{P}\Big[A'(1 - e^{-Pt}) + \eta t\Big] \quad \text{Eq 43}$$

where, $A' = 1-(\eta/P)$, Eq 44

And for the decay products:

$$N_A = \frac{N_1^0 T\lambda_n}{P}\left[C + \frac{\eta}{\lambda_A}t - \left(\frac{A'}{\lambda_A - P}\right)e^{-Pt} - Be^{-\lambda_A t}\right] \quad \text{Eq 45}$$

where, $B = \left(\frac{A'}{\lambda_A} - \frac{\eta}{\lambda_A^2}\right) - \frac{A'}{\lambda_A - P}$, and Eq 46

$$C = \left(\frac{A'}{\lambda_A} - \frac{\eta}{\lambda_A^2}\right) \quad \text{Eq 47}$$

$$N_B = \frac{N_1^0 T\lambda_n\lambda_A}{P}\left[D + \frac{\eta}{\lambda_A\lambda_B}t - Fe^{-Pt} - \frac{B}{\lambda_B - \lambda_A}e^{-\lambda_A t} - Xe^{-\lambda_B t}\right] \quad \text{Eq 48}$$

where, $$D = \left(\frac{C}{\lambda_B} - \frac{\eta}{\lambda_A \lambda_B^2}\right) \quad , \qquad \text{Eq 49}$$

$$F = \frac{A'}{(\lambda_A - P)(\lambda_B - P)} \quad , \text{ and} \qquad \text{Eq 50}$$

$$X = D - F - \frac{B}{\lambda_B - \lambda_A} \qquad \text{Eq 51}$$

$$N_C = \frac{N_1^0 T \lambda_n \lambda_A \lambda_B}{P}\left[G + Ht - Ie^{-Pt} - Je^{-\lambda_A t} - Le^{-\lambda_B t} - Re^{-\lambda_C t}\right] \qquad \text{Eq 52}$$

where, $$G = \left(\frac{D}{\lambda_C} - \frac{\eta}{\lambda_A \lambda_B \lambda_C^2}\right) \quad , \qquad \text{Eq 53}$$

$$H = \frac{\eta}{\lambda_A \lambda_B \lambda_C} \qquad \text{Eq 54}$$

$$I = \frac{F}{\lambda_C - P} \qquad \text{Eq 55}$$

$$J = \frac{B}{(\lambda_B - \lambda_A)(\lambda_C - \lambda_A)} \qquad \text{Eq 56}$$

$$L = \frac{X}{\lambda_C - \lambda_B} \qquad \text{Eq 57}$$

$$R = G - (I + J + L) \qquad \text{Eq 58}$$

$$N_{C'} = 0.663\, N_C(\lambda_C/\lambda_{C'}) \qquad \text{Eq 59}$$

The integrated activities are calculated using Equations 12 and 13, and Equations 43, 45, 48 and 52. Hence:

$$IAS(n) = \frac{N_1^0 T \lambda_n}{P}\left[A'(T_2 - T_1) + \frac{A'}{P}\left(e^{-PT_2} - e^{-PT_1}\right) + \frac{\eta}{2}(T_2^2 - T_1^2)\right] \qquad \text{Eq 60}$$

$$IAS(A) = \frac{N_1^0 T \lambda_n \lambda_A}{P}\left[C(T_2 - T_1) + \frac{\eta}{2\lambda_A}(T_2^2 - T_1^2) + \frac{A'}{P(\lambda_A - P)}\left(e^{-PT_2} - e^{-PT_1}\right) + \frac{B}{\lambda_A}\left(e^{-\lambda_A T_2} - e^{-\lambda_A T_1}\right)\right] \qquad \text{Eq 61}$$

$$IAS(B) = \frac{N_1^0 T \lambda_n \lambda_A \lambda_B}{P}\left[D(T_2 - T_1) + \frac{\eta}{2\lambda_A \lambda_B}(T_2^2 - T_1^2) + \frac{F}{P}\left(e^{-PT_2} - e^{-PT_1}\right) + \frac{B}{\lambda_A(\lambda_B - \lambda_A)}\left(e^{-\lambda_A T_2} - e^{-\lambda_A T_1}\right) + \frac{X}{\lambda_B}\left(e^{-\lambda_B T_2} - e^{-\lambda_B T_1}\right)\right] \qquad \text{Eq 62}$$

$$IAS(C) = \frac{N_1^0 T \lambda_n \lambda_A \lambda_B \lambda_C}{P} \left[G(T_2 - T_1) + \frac{H}{2} (T_2^2 - T_1^2) \right.$$

$$+ \frac{I}{P} \left(e^{-PT_2} - e^{-PT_1} \right) + \frac{J}{\lambda_A} \left(e^{-\lambda_A T_2} - e^{-\lambda_A T_1} \right)$$

$$\left. + \frac{L}{\lambda_B} \left(e^{-\lambda_B T_2} - e^{-\lambda_B T_1} \right) + \frac{R}{\lambda_C} \left(e^{-\lambda_C T_2} - e^{-\lambda_C T_1} \right) \right] \qquad \text{Eq 63}$$

and, $$IAS(C') = 0.663\ IAS(C) \qquad \text{Eq 64}$$

1c. EXPONENTIAL CASE

In this case, Equation 5 becomes:

$$N_1 = N_1^0\, e^{\delta t} \qquad \text{Eq 65}$$

which, upon substitution into Equation 4, taking into account the proper boundary condition, gives:

$$N_n = \frac{N_1^0 T}{\delta + P} \left(e^{\delta t} - e^{-Pt} \right) \qquad \text{Eq 66}$$

$$N_A = \frac{N_1^0 T \lambda_n}{\delta + P} \left(\frac{e^{\delta t}}{\delta + \lambda_A} - \frac{e^{-Pt}}{\lambda_A - P} + \phi e^{-\lambda_A t} \right) \qquad \text{Eq 67}$$

where, $$\phi = \frac{P + \delta}{(\delta + \lambda_A)(\lambda_A - P)} \qquad \text{Eq 68}$$

$$N_B = \frac{N_1^0 T \lambda_n \lambda_A}{\delta + P} \left[\frac{e^{\delta t}}{(\delta + \lambda_A)(\delta + \lambda_B)} - \frac{e^{-Pt}}{(\lambda_A - P)(\lambda_B - P)} \right.$$

$$\left. + \frac{\phi}{\lambda_B - \lambda_A} e^{-\lambda_A t} - \Omega e^{-\lambda_B t} \right] \qquad \text{Eq 69}$$

where, $$\Omega = \frac{1}{(\delta + \lambda_A)(\delta + \lambda_B)} - \frac{1}{(\lambda_A - P)(\lambda_B - P)} + \frac{\phi}{\lambda_B - \lambda_A} \qquad \text{Eq 70}$$

$$N_C = \frac{N_1^0 T \lambda_n \lambda_A \lambda_B}{\delta + P} \left(m e^{\delta t} - n e^{-Pt} + q e^{-\lambda_A t} - r e^{-\lambda_B t} - \psi e^{-\lambda_C t} \right) \qquad \text{Eq 71}$$

where, $$m = \frac{1}{(\delta + \lambda_A)(\delta + \lambda_B)(\delta + \lambda_C)} \qquad \text{Eq 72}$$

$$n = \frac{1}{(\lambda_A - P)(\lambda_B - P)(\lambda_C - P)} \qquad \text{Eq 73}$$

$$q = \frac{\phi}{(\lambda_B - \lambda_A)(\lambda_C - \lambda_A)} \qquad \text{Eq 74}$$

$$r = \frac{\Omega}{\lambda_C - \lambda_B} \quad , \text{ and} \qquad \text{Eq 75}$$

$$\psi = m - n + q - r \qquad \text{Eq 76}$$

$$N_{C'} = 0.663\ N_C(\lambda_C/\lambda_{C'}) \qquad \text{Eq 77}$$

The integrated activities are calculated using Equations 12, 13 and Equations 66, 67, 69 and 71. Hence:

$$IAS(n) = \frac{N_1^0 T \lambda_n}{\delta + P}\left[\frac{1}{\delta}\left(e^{\delta T_2} - e^{\delta T_1}\right) + \frac{1}{P}\left(e^{-PT_2} - e^{-PT_1}\right)\right]$$

$$IAS(A) = \frac{N_1^0 T \lambda_n \lambda_A}{\delta + P}\left[\frac{e^{\delta T_2} - e^{\delta T_1}}{\delta(\delta + \lambda_A)} + \frac{e^{-PT_2} - e^{-PT_1}}{P(\lambda_A - P)} - \frac{\phi}{\lambda_A}\left(e^{-\lambda_A T_2} - e^{-\lambda_A T_1}\right)\right] \qquad \text{Eq 79}$$

$$IAS(B) = \frac{N_1^0 T \lambda_n \lambda_A \lambda_B}{\delta + P}\left[\frac{e^{\delta T_2} - e^{\delta T_1}}{\delta(\delta + \lambda_A)(\delta + \lambda_B)} + \frac{e^{-PT_2} - e^{-PT_1}}{P(\lambda_A - P)(\lambda_B - P)}\right.$$

$$\left. - \frac{\phi}{\lambda_A(\lambda_B - \lambda_A)}\left(e^{-\lambda_A T_2} - e^{-\lambda_A T_1}\right) + \frac{\Omega}{\lambda_B}\left(e^{-\lambda_B T_2} - e^{-\lambda_B T_1}\right)\right] \qquad \text{Eq 80}$$

$$IAS(C) = \frac{N_1^0 T \lambda_n \lambda_A \lambda_B \lambda_C}{\delta + P}\left[\frac{m}{\delta}\left(e^{\delta T_2} - e^{\delta T_1}\right) + \frac{n}{P}\left(e^{-PT_2} - e^{-PT_1}\right)\right.$$

$$- \frac{q}{\lambda_A}\left(e^{-\lambda_A T_2} - e^{-\lambda_A T_1}\right) + \frac{r}{\lambda_B}\left(e^{-\lambda_B T_2} - e^{-\lambda_B T_1}\right)$$

$$\left. + \frac{\psi}{\lambda_C}\left(e^{-\lambda_C T_2} - e^{-\lambda_C T_1}\right)\right] \qquad \text{Eq 81}$$

and, $IAS(C') = 0.663\ IAS(C)$

2. REVERSE DIFFUSION PROCESS

Solutions for the radioactive gas atom concentration and its progeny atom concentration for the reverse diffusion process are obtained by using Equation 10 with the following boundary conditions:

$$N_n(T_w = 0) = N_n(T_s)$$

$$N_A(T_w = 0) = N_A(T_s)$$

$$N_B(T_w = 0) = N_B(T_s), \text{ and}$$

$$N_C(T_w = 0) = N_C(T_s)$$

where, $N_n(T_s)$, $N_A(T_S)$, $N_B(T_s)$ and $N_C(T_s)$ are the atom concentrations at the

end of the direct diffusion process. These atom concentrations are obtained by substituting t by T_s in Equations 15, 16, 21, 25, 43, 45, 48, 52, 66, 67, 69, and 71.

Solution of Equation 10, taking into account the above boundary conditions, gives the atom concentration for the reverse diffusion process in the time-dependent direct diffusion case. (It should be noted that for the radioactive gas, the first term on the right hand side of Equation 10 should be taken as zero.) The expressions for the activities and integrated activities are then obtained using Equations 11, 12 and 13 as before.

The procedure outlined above is identical to the one followed for the steady-state reverse diffusion case, as discussed elsewhere (7). Hence, one may use Equations 19, 20, 21 and 22 of ref. (7), taking into account that $N_n(T_s)$, $N_A(T_s)$ and so on, are this time those corresponding to the time-dependent direct diffusion process, in order to obtain N_n, N_A, and so on, and IAW(n), IAW(A), and so on for the reverse diffusion process, under time-dependent conditions.

If for practical reasons an α-detector is used in the sensitive volume, the total α-count recorded by the detector and associated electronic circuitry is given by Equation 28 (ref. 7). A similar expression, although somewhat reduced, applies for the case of a β-detector to obtain total β-count.

REMARKS

The expressions derived in the present work and in a previous paper have been used extensively to study the dynamic behaviour of diffusion and radon and thoron through membranes under external (radiation) steady-state conditions and time-dependent conditions. This work finds application to personal dosimetry and environmental monitoring by passive diffusion. The

work is also of interest from the theoretical standpoint and can find, in addition, a variety of applications of practical interest.

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